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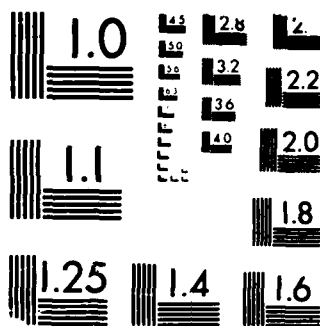
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# LIGHT SCATTERING BY CHARGED SPHERES

PREPARED BY  
ALESSANDRO COLETTI

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19. ABSTRACT (Continue on reverse if necessary and identify by block number) The major goal of this project was to verify experimentally the theoretical model that Bohren and Hunt (1977) proposed to describe light scattering by electrically charged spherical particles. Our experimental set-up did not allow us to totally verify their model because of the preliminary nature of our measurements. However the experimental results obtained can be considered as evidence where experimental changes in the light scattering by spherical particles can be ascribed to surface charge effects.			
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LIGHT SCATTERING BY CHARGED SPHERES

FINAL REPORT

ALESSANDRO COLETTI

JANUARY 1988

U.S. ARMY RESEARCH OFFICE

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## 1. Introduction

The Lorenz-Mie theory for light scattering by homogeneous isotropic spheres assumes that the spheres are uncharged and that there are no external electric fields. The wide range of validity of the Lorenz-Mie theory has been assessed in the laboratory and in the applications; therefore the experimental errors in measurements of particle light scattering have never been associated with surface charge effect because of the unknown relevance of the problem. However, the presently growing interest on electrooptical effects and non-linear optics, justified this short investigation aimed to verify directly the effect of surface charges on particle light scattering.

Direct evidence of optical effects of surface charges has been obtained by Feinlab (1966) using synchronous light detection on polished gold electrodes in an electrolytic bath configuration. Garrigos et al (1974) interpreted Feinlab's results with a model of surface conductivity on metallic gold and Bohren and Hunt have extended the Lorenz-Mie theory to the case of charged spherical particles by including a surface conductivity term in the calculations.

The surface conductivity can be visualized as a monomolecular layer having optical properties different from the bulk of the material whose properties can be represented by a complex number. The real part of the surface conductivity accounts for the electrical losses of the incoming wave on the surface, the imaginary part for its electrical impedance.

In this note we present experimental results where the light scattering by glass particles systematically varied with their surface charge, and some numerical results describing the effect on spherical particle of the surface conductivity according to the Bohren and Hunt model.

Even though at the present time theoretical treatments analogous to the one presented here for the spherical case are not available for cylinders, or for other non-spherical geometries, nevertheless our numerical results can give some indication to what can be expected in the other cases.

The experiments conducted in our laboratory on glass spheres in the electrostatic case, have shown a variation of the light scattering with the surface charge of the particles. Since our experimental set up did not allow us to exclude conclusively the presence of possible undesired experimental effects, we do not consider these experiments conclusive.

## 2. The Theoretical Model

The Bohren and Hunt model can be summarized considering a particle, with radius  $a$ , charged with charge  $Q$  confined onto its surface. Then it is possible to define an excess surface charge density,  $\eta_s$ , on the surface  $A$  of the particle so that:

$$\int \eta_s dA = Q \quad 1)$$

When the surface charge is acted upon an oscillatory electric field, it is useful to divide the charge density into two components, a static component  $\eta_0$ , and an oscillatory component  $\eta_e$ , so that the total charge can be conserved in the calculations:

$$\eta_s = \eta_0 + \eta \quad 2)$$

where

$$\eta = \text{Re} \{ \tilde{\eta} \exp(-i\omega t) \} \quad 3)$$

and

$$\int \eta_0 dA = Q \quad 4)$$

$$\int \tilde{\eta} dA = 0 \quad 5)$$

At this point a surface current density can be defined introducing the vector  $u_t$  of the tangential velocity of the charge on the surface:

$$K = \eta_s u_t \quad 6)$$

and as a consequence a surface conductivity  $\sigma_s$  can be introduced:

$$K = \sigma_s E_{1t} \quad 7)$$

The surface conductivity of the SI units can be expressed in  $(\text{ohm m})^{-1}$  and can be thought of as an empirical complex quantity characteristic of the surface of the material under consideration at the frequency of the field.

This equation is used by Bohren and Hunt to impose the boundary condition on the field in the medium that represents the superimposition of the incident and of the scattered field.

Denoting with subscript 1 the electric and magnetic fields inside the particle and with subscript 2 the fields outside, the system of the boundary condition on the surface of a particle of radius  $a$ , can be expressed as:



$$\mathbf{n} \times (\mathbf{E}_2 - \mathbf{E}_1) = 0$$

8)

$$\mathbf{n} \times (\mathbf{H}_2 - \mathbf{H}_1) = \mathbf{K}$$

so that using equation 7, it results:

$$\mathbf{n} \times (\mathbf{H}_2 - \mathbf{H}_1) = \sigma_s \mathbf{E}_{1t} \quad 9)$$

Within this hypothesis Bohren and Hunt developed their final results for the light scattering coefficients of the sphere where as usual:

$$C_{sca} = \left(\frac{2\pi}{km}\right)^2 \sum_{n=1}^{\infty} (2n+1) (|a_n|^2 + |b_n|^2) \quad 10)$$

and

$$a_n = -A_n/V_n ; b_n = -B_n/W_n \quad 11)$$

but

$$\begin{aligned} A_n &= \psi_n(mx) \psi_n'(x) - m \psi_n'(mx) \psi_n(x) + i \omega \sigma_s \mu_0 \psi_n(x) \psi_n(mx) / k_m \\ V_n &= \psi_n(mx) \xi_n'(x) - m \psi_n'(mx) \xi_n(x) + i \omega \sigma_s \mu_0 \psi_n(mx) \xi_n(x) / k_m \\ B_n &= \psi_n'(mx) \psi_n(x) - m \psi_n'(x) \psi_n(mx) - i \omega \sigma_s \mu_0 \psi_n'(x) \psi_n'(mx) / k_m \\ W_n &= \psi_n'(mx) \xi_n(x) - m \psi_n(mx) \xi_n'(x) - i \omega \sigma_s \mu_0 \psi_n'(mx) \xi_n'(x) / k_m \end{aligned} \quad 12)$$

contains new additional terms that do not appear in the Lorenz-Mie theory.

To understand the effect of the extra terms in the numerator and denominator of equation 11, it is useful to look at the scattering resonances discussed by Chylek et al (1978) for the spherical case.

For our calculations we have modified a light scattering program based on Wiscombe numerical method, and we have introduced the additional terms required by the equations.

The differences in the scattering coefficient existing between a spherical

particle with a refractive index  $n=1.5$  and a similar particle with real surface charge conductivity equal to  $5 \times 10^{-8}$  SI units are shown in Figure 1. The figure shows how the introduction of a real surface conductivity decreases the scattering coefficient in the overall and at the same time dumps the amplitude of the natural resonances of the sphere. In fact, a purely real surface conductivity is equivalent to a purely resistive impedance on the surface that absorbs the incoming field on a non-light absorbing particle. Since this light absorption is occurring on the particle surface and in the majority of the natural processes involving evaporation or condensation the total charge is conserved, we can expect that the absorption coefficient of charged particle increases, decreasing the particle size parameter. For instance, Figure 2 compares extinction, scattering and absorption coefficients of two particles one light absorbing with a refractive index  $n=1.5 - i 0.005$  and one with a refractive index purely real and a surface conductivity of  $5 \times 10^{-7}$ . The three coefficients assume approximately the same values for both particles at a size of about 16, but as expected, while the absorption coefficient of the light absorbing particle increases with the size parameter that of the charged particle decreases after reaching a maximum at about size 2.3.

Figure 3 compares the scattering coefficients of a non-light absorbing particle with that of charged particles for different values of the imaginary part of the surface conductivity (real part insignificantly small). Basically the imaginary surface conductivity affects the position of the resonant peaks of the sphere without changing significantly the average value of the scattering coefficient. Therefore, the effect of an imaginary component of the surface conductivity can be of interest in precision measurements of particle sizes or in micro particle Raman spectroscopy where the position of the natural resonances of a sphere are used to determine its size or its chemical composition (Arnold, 1980; Lettieri, 1984).

High resolution plots of light scattering of single particles reveal the effect of the surface charges that otherwise is insignificant on integrated quantities. In fact in the case of interstellar particle polydispersions, Professor Giese's group (Weiss, 1981) following the Bohren and Hunt calculations, estimated a correction in the light scattering of the order of one part over in  $10^3$ .

Furthermore, similar conclusions are also justified when looking at particle phase functions. In fact, the clearly measurable variation of the phase function reported in Figure 4 corresponds to a surface conductivity value

ten times larger than the one shown in the previous figures ( $5 \times 10^{-7}$  against  $5 \times 10^{-6}$  in Figure 3), and such high values can be considered probably unlikely to occur.

### 3. The Experimental Method

For the experiments we have used an electrostatic particle levitation system that allows estimates of the particle charges and determinations of the light scattering resonances (Lettieri and Jenkins, 1984).

Our experimental set-up consisted of a tunable dye laser to illuminate the particle, an electrostatic levitation system for particle suspension, a microscope to stabilize visually the position of the particle within the electric field, and a monochromator tuned on the laser line.

The electrostatic levitation system served the function to keep the particle suspended in the center of the system, to measure the particle charges, and to measure the particle levitation force under the action of the laser. The system has been built and tested with the generous help of Dr. D. Capps (Boeing Co.) who also gave us very helpful suggestions during the program. We also kept close scientific contact with Dr. Arnold of the Polytechnic Institute of New York, and with Professor Bohren for the theoretical interpretation of the data.

To obtain charged particles there are various methods. Metallic particles, electrostatically suspended inside a quadrupole, can be charged with ultraviolet light before being conductive liquid solutions or suspensions can be charged keeping electrical contact between the bulk of the liquid in the spraying system and a power supply that polarizes a metallic ring placed in front of the nozzle (Liu and Pui, 1974).

In our experiments we have used solid glass spheres 15mm diameter, and a random amount of surface charge was simply obtained by triboelectric electrification. The polarization potential necessary to stabilize the particle in the quadrupole was used to quickly estimate if the particle captured in the field had a charge amount sufficient for staying balanced in the field after being discharged a few times with our radioactive source. Typically the charge amount that was achieved during the measurements with this method was of the order of  $10^{-5} \text{ C m}^{-2}$  (equivalent to  $10^{16}$  electrons on the particle surface). Therefore as far as particle charging, our experiments can be considered to have been made under similar conditions as those performed by Bottiger et al (1980), and Wyatt and Phillips (1972).

During the experiments we have measured the light scattered at  $90^\circ$  by the

glass particles at 15 different wavelengths in the interval between 0.6570 and 0.6530  $\mu$ m. The signal of the photomultiplier was constantly compared with the power emitted by the laser averaging the data in a multichannel analyzer.

Figure 5 shows a typical plot of the measurements. Each set of experimental points has been obtained for constant value of the particle surface charge and changing the wavelength of the dye laser. From the figure it appears that there are differences in the light scattering of the glass particle at the various levels of surface charge, that are well above the experimental errors. These variations cannot be accounted for by accidental variations of the luminous power of the dye laser. Under these circumstances it would appear that if the measured effect is actually due to the surface charge effect, the surface conductivity of our glass spheres was of the same order of magnitude of the one used in Figure 3 of  $10^{-5}$ . This effect is rather unexpected at the low levels of surface charge densities used during the experiments, and unfortunately we have not been able to perform further investigations under vacuum in order to be able to exclude any contamination on the particle surface, and recharging the particles with ultraviolet light, to verify that, their light scattering coefficient would actually assume the values obtained before the discharge.

#### 4. Conclusions

Variations of the light scattering by single electrostatically levitated spheres have been observed when changing the amount of the surface charge. The amount of the surface charges on our sample spheres was changed by exposing a radioactive source, located in the proximity of the sphere, at the end of each scan in wavelength. Before and after each experiment particular attention was spent in verifying that the spheres under use were regular and homogeneous under the optical microscope. This was done to insure that there were no visible deposits on the surface of the sample particle. However, since the basic experimental set-up was such that it was taking about 10 minutes to perform 15 readings at different wavelengths, and at times the particle surface charge would slightly change because of background natural radioactivity, we have not been able to obtain more than one scan in wavelength under the same experimental conditions.

Therefore, in our opinion, these experiments proved only that under laboratory controlled conditions, sensible optical effect can be observed on Mie spheres by subtle changes of the properties of the surface. The conclusion of this research period is that slight changes in the optical properties of glass

spheres can occur in time or varying the surface charge. Unfortunately, a separation of the two effects has not been possible with our experimental set-up. However, methods closer to those realized by Feinlab, where time varying potentials were used along with synchronous detection, might permit to separate the effect of surface charges versus that of possible surface contamination.

Our measurements, even within their limitations, have proven that the optical properties of surface can be seriously affected even by inconspicuous factors so that the difficulties experienced in measuring in the IR the optical constants of powders not only is not surprising but also might be originated by factors whose nature could be much more complex than generally believed. Within this perspective our measurements can be considered a first attempt toward the process of defining experiments for the direct investigations on isolated particle on these possible factors.

5. List of Participating Personnel

Dr. A. S. Coletti (PI)

Mr. C. M. Wyman

Graduate Student: Mr. Alan Chu

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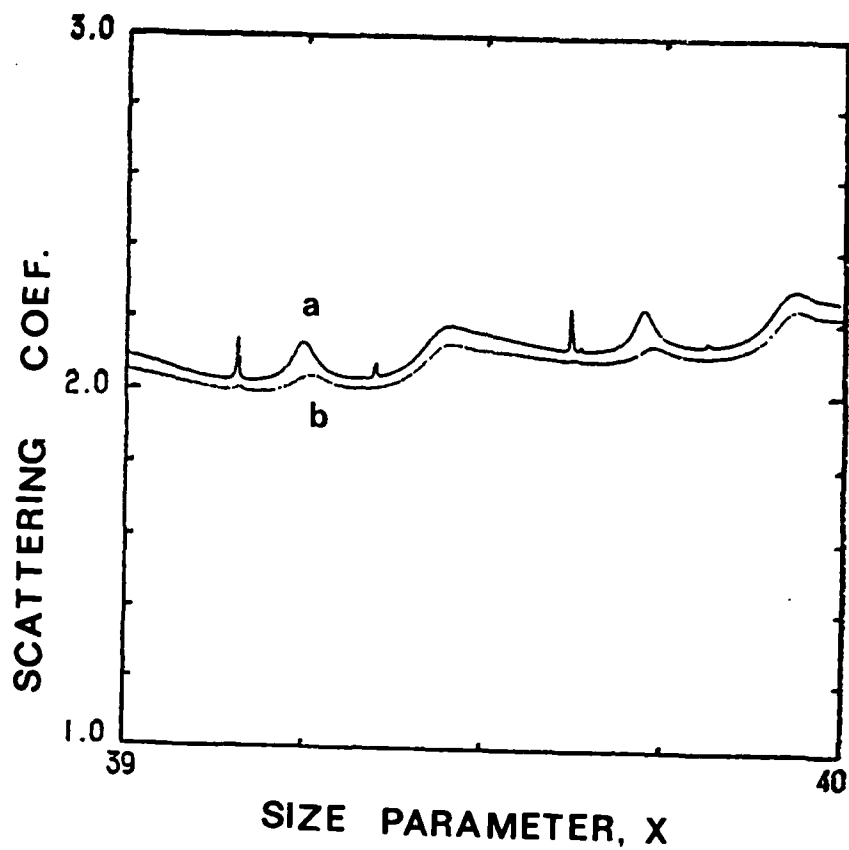


Fig.1 Effect of the real part of the surface conductivity on the scattering coefficient as function of the size parameter (refractive index 1.5). a) surface conductivity 0; b) surface conductivity  $5 \times 10^{-8}$ .

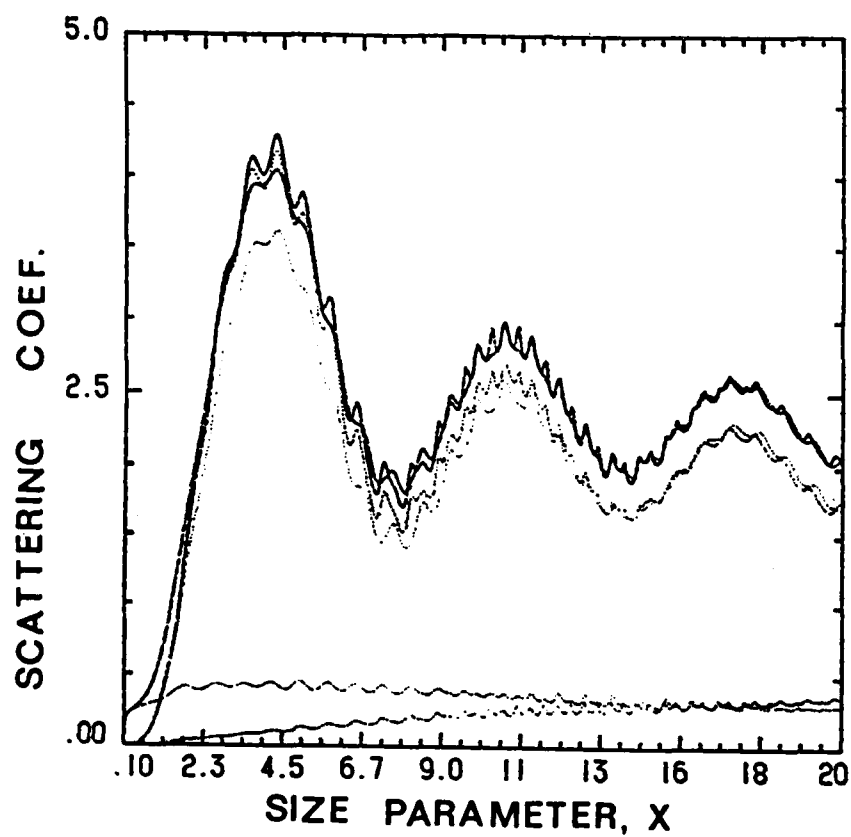


Fig. 2 Extinction, scattering, and absorption coefficient of charged spheres with surface conductivity  $5 \times 10^{-7}$  ( $m = 1.5$ ), and of light absorbing spheres ( $m = 1.5 - i 0.005$ )



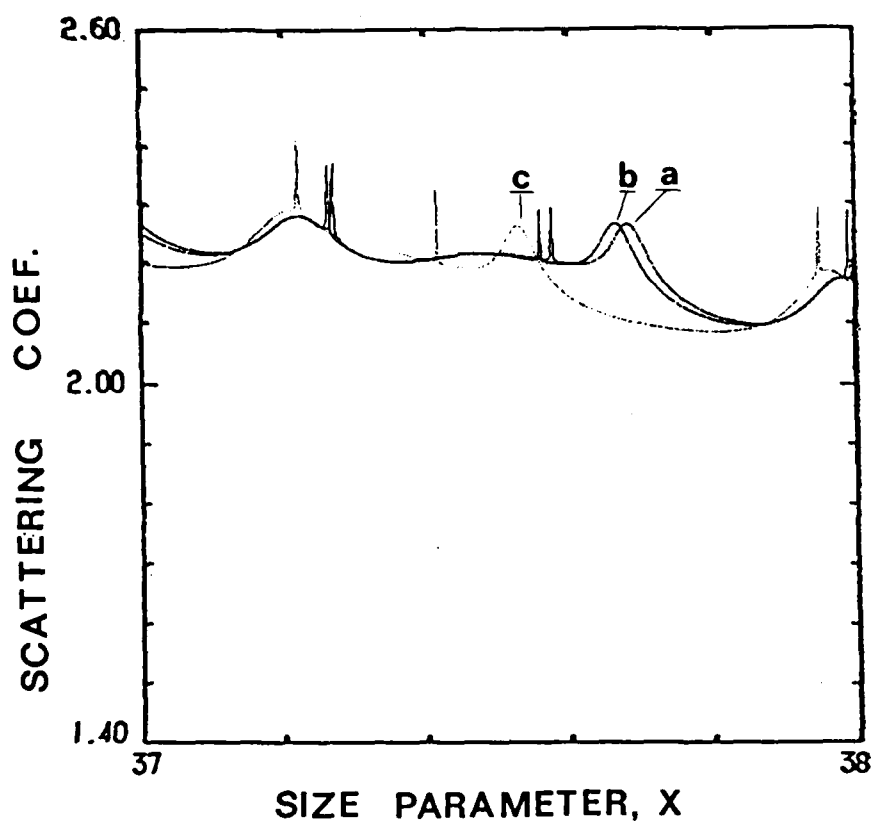


Fig.3 Effect of the imaginary part of the surface conductivity on the scattering coefficient as function of the size parameter (refractive index 1.5). a) surface conductivity 0, b) surface conductivity  $-5 \times 10^{-8}i$ , c) surface conductivity  $-5 \times 10^{-7}$ .

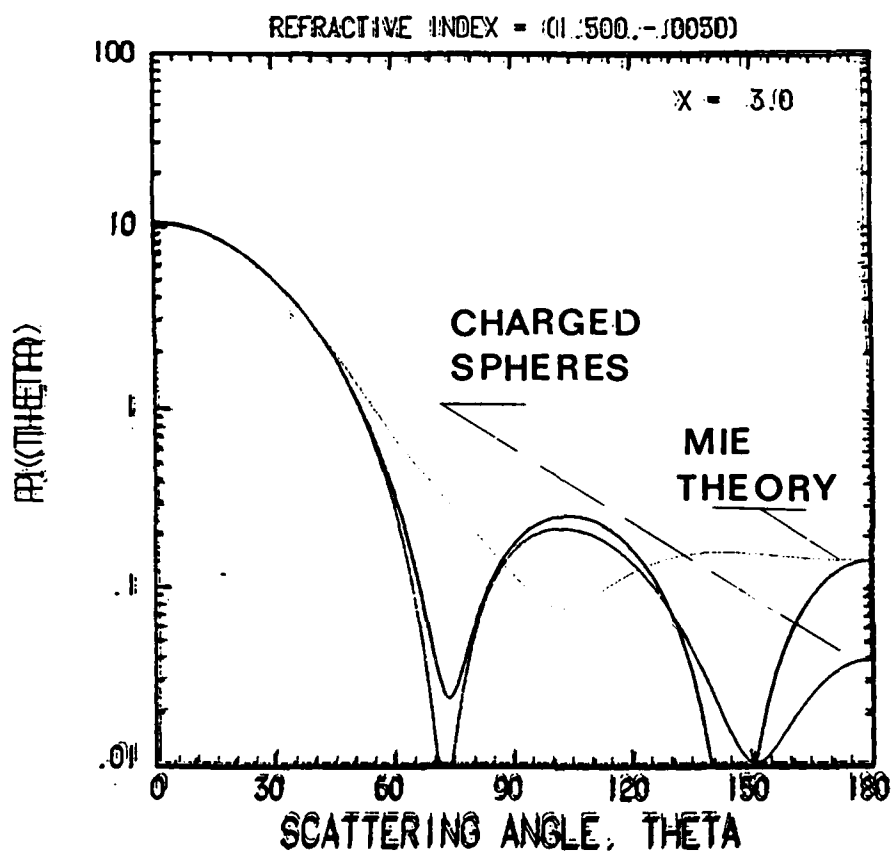


Fig.4 Polarized light scattering phase functions for a particle of size parameter  $x=3$ , refractive index 1.5, in the case of no surface charge and in the case of a surface conductivity of  $5 \times 10^{-7}$ .

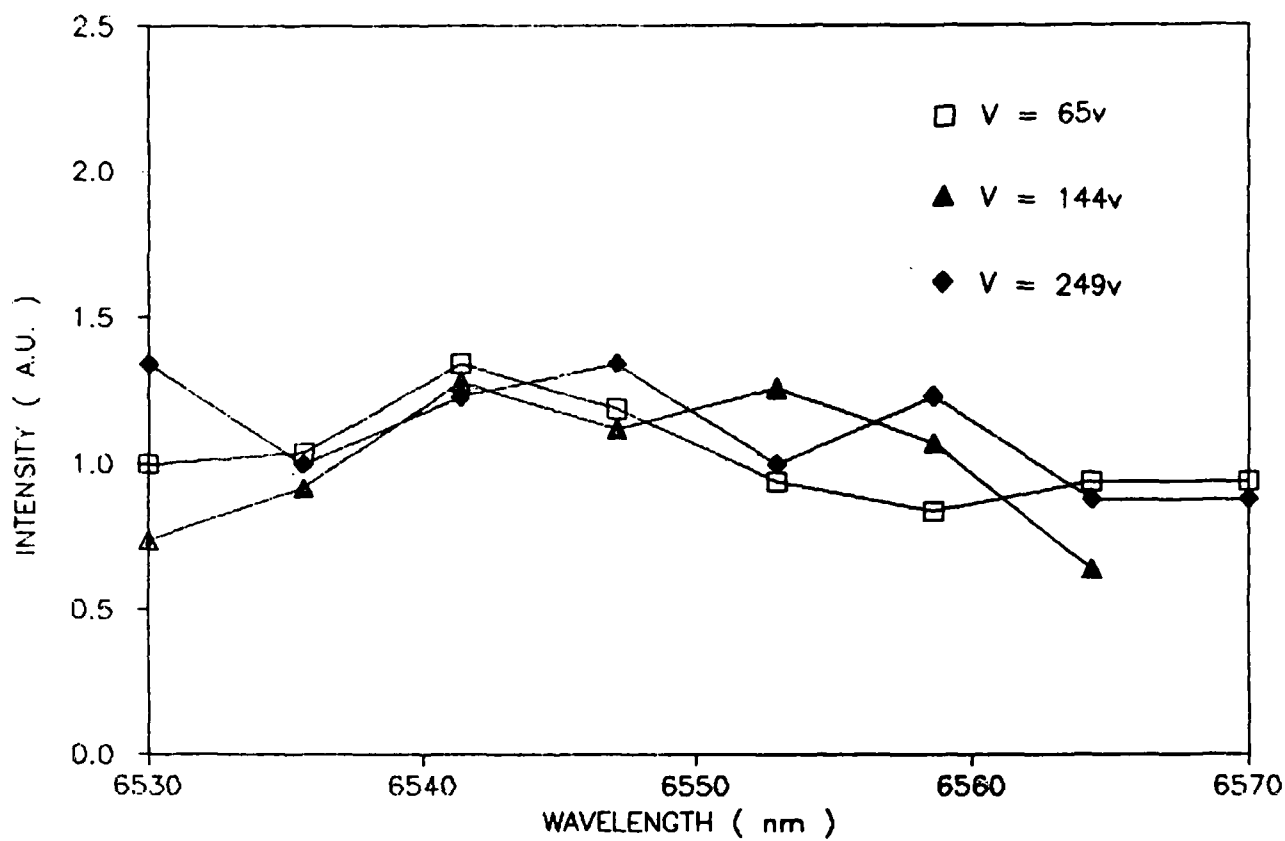


Figure 5 Measured light scattering at  $90^\circ$  of glass spheres for various values of the levitating voltage as function of the dye laser wavelength.

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